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Bismuth-210, its parent, and daughter and their use as particle tracers in aquatic systems

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ABSTRACT

Disequilibrium between ²¹⁰Pb (t½: 22.3 years) and its granddaughter ²¹⁰Po (t½: 138.4 days) has been used to infer organic particle removal rates from the surface ocean on a timescale of weeks to months; however, the behavior of the short-lived intermediary daughter ²¹⁰Bi (t½: 5.01 days) is not well known. Here, timeseries measurements of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in the Milwaukee Inner Harbor on Lake Michigan showed respective particle distribution (K_d) coefficients of 7.1 (± 0.4) × 10⁵, 9.3 (± 1.7) × 10⁵, and 10.2 (± 0.6) × 10⁵ L kg⁻¹, giving K_d ratios, or fractionation factors (F), of $F_{Bi/Pb} = 1.3 \pm 0.3$, $F_{Po/Bi} = 1.1 \pm 0.2$, and $F_{Po/Pb}$ of 1.4 ± 0.1 . After a large rain event, total ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activities in the harbor water column decreased over a course of 12 days from 347 to 105 dpm ²¹⁰Pb m⁻³, 397 to 88 dpm ²¹⁰Bi m⁻³, and 144 to 58 dpm ²¹⁰Po m⁻³. This is the first observation of a total ²¹⁰Bi/²¹⁰Po activity ratio of <1 in an aquatic system. A 1-D non-steady-state model of nuclide fluxes suggested that 92% of the reduction in ²¹⁰Bi activity was due to removal from the water column, with the remainder accounted for by net in situ decay. The potential for using ²¹⁰Bi as a tracer of high and variable particle scavenging rates in lakes and shallow marine systems is supported.

1. Introduction

Chemical cycling in any aquatic system is significantly mediated by the transport of particles, and a better understanding of particle dynamics is particularly needed in lakes and shallow marine systems (e.g., Hanson et al., 2015; Pauer et al., 2011; Bohlen et al., 2012; Cael and Bisson, 2018), which play a disproportionately large, yet poorly quantified role in global biogeochemical cycling (e.g., Dean and Gorham, 1998; Muller-Karger et al., 2005; Fennel et al., 2006; Giraud et al., 2008; Drake et al., 2018). Our current understanding of particle flux and particle attenuation down through the water column has largely relied on the use of sediment traps and naturally occurring particle-reactive radionuclides. However, the downward transport of particulate matter in shallow waters cannot be reliably quantified with a bottom tethered sediment trap (e.g., Boyce et al., 1990; Buesseler et al., 2007; Buesseler et al., 2010; Waples, 2015) and the choice of radionuclide tracers in lowsalinity and high flux systems is severely limited.

One radiotracer candidate for low-salinity systems is the naturally occurring radionuclide lead-210 (210 Pb; t/₂: 22.3 years). 210 Pb is near the end of the 238 U decay chain and is found in all aquatic systems by means of in situ decay of its parents or through atmospheric deposition from the

decay of ²²²Rn gas that has leaked from the continental crust. ²¹⁰Pb and its granddaughter polonium-210 (²¹⁰Po; t¹/₂: 138.4 days) have long been used as a proxy for particle flux from the surface ocean, where the preferential sorption of ²¹⁰Po to sinking particles and the resultant disequilibrium between ²¹⁰Pb and ²¹⁰Po can be used to infer net particle removal rates on a timescale of weeks to months (e.g., Shannon et al., 1970; Bacon et al., 1976; Rutgers van der Loeff and Geibert, 2008; Verdeny et al., 2009). However, the timescale of this Po/Pb tracer pair is inappropriate for shallow and high flux environments.

 210 Pb decays to 210 Po through the short-lived intermediary daughter bismuth-210 (210 Bi; t½: 5.01 days). Biggin et al. (2002) and Fowler et al. (2010) have suggested that 210 Bi might prove useful as a tracer for fast scavenging processes, where rapid ingrowth of 210 Bi can quickly respond to both high and variable particle scavenging rates on a timescale of hours to days; however, little is known about this nuclide's behavior in the water column.

The use of 210 Bi as a particle tracer depends foremost on bismuth's affinity for particles. Bismuth is a group 15 element with an oxidation state of +3. Its principal species in both freshwater and marine systems (pH 6–9) is Bi(OH)₃ (Turner et al., 1981; Byrne, 2002), and binding of bismuth to oxyhydroxides of iron and manganese is evidenced by its

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high concentrations in Fe and Mn minerals (Bertine et al., 1996; Fowler et al., 2010). Lee's (1982) vertical profile of [nearly] stable ²⁰⁹Bi in the north Pacific Ocean mirrored that of manganese and clearly showed scavenging in the mixed surface and bottom layers of the water column with up to 70% of the bismuth partitioned to the particulate fraction. ²¹⁰Bi should behave like its stable isotope; however, the partitioning of ²¹⁰Bi between dissolved and particulate fractions in a natural system has only been measured once – in a single sample from the Irish Sea (Biggin et al., 2002).

The goal of this study was to better understand how ²¹⁰Bi behaves in a natural aquatic system. In a previous paper (Waples, 2020), I described a method for quickly measuring ²¹⁰Bi, its parent ²¹⁰Pb, and daughter ²¹⁰Po; and provided – as proof of concept – activity measurements of all three nuclides in water samples collected from the Milwaukee Inner Harbor on Lake Michigan. In this paper, I elaborate on these same activity measurements in conjunction with measurements of suspended particulate matter to determine particle distribution coefficients for all three nuclides. I then use a one-dimensional (1-D) non-steady-state (NSS) model to demonstrate the calculation of nuclide and particulate matter fluxes over the sampling time course. These observations are then used to assess the use of ²¹⁰Bi as a tracer for rapid scavenging processes.

2. Materials and methods

2.1. Study site

Samples of rain and lake water were collected from the University of Wisconsin-Milwaukee School of Freshwater Sciences (SFS) campus and its adjacent slip (average depth: \sim 4 m) in the Milwaukee Inner Harbor (Fig. 1).

2.2. Sample collection

Rainwater was collected at ground level over a 40-h period during an



Fig. 1. Study area in the Milwaukee Inner Harbor at the University of Wisconsin-Milwaukee School of Freshwater Sciences (circle). Rainwater collected at 43.017719° N, 87.903000° W; lake water collected at 43.017835° N, 87.903521° W.

extended rain event beginning at 17:20 on 03 November 2018. Lake water samples (~50 L) were collected from the slip with a submersible pump from a depth of 2 m on 08, 15, and 20 November 2018. An additional water sample (~20 L) was collected during each sampling event for the determination of suspended particulate matter (SPM; > 0.45 μ m).

2.3. Radionuclide analysis

A detailed description of the methods for determining in situ ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activities in rain and lake water is given by Waples (2020). Briefly, lake water samples were weighed and filtered through nitrocellulose filters (0.45 µm, 293 mm, Millipore) within two hours of sample collection to separate dissolved and particle-bound nuclide fractions. Yield monitors ²⁰⁷Bi and ²⁰⁹Po (Eckert & Ziegler Isotope Products) were then added to both sample fractions. Dissolved nuclide fractions (< 0.45 μ m) of ²¹⁰Bi, and ²¹⁰Po were co-precipitated onto newly formed ferric hydroxide and collected by filtration. Both particlebound and dissolved nuclide fractions were subsequently digested in concentrated nitric acid. Total ²¹⁰Pb was determined in a separate water sample via ²¹⁰Bi that had grown into secular equilibrium with its parent. Bismuth and polonium were completely separated from lead on EmporeTM anion solid phase extraction (SPE) disks (3 M, 47 mm, product number 2252, now manufactured by CDS Analytical). Eluate from the particle-bound fraction was re-spiked with 207 Bi and set aside for ~one month for the determination of particle-bound ²¹⁰Pb (via ²¹⁰Bi in secular equilibrium with its parent). Anion SPE disks were immediately counted - first for ²¹⁰Bi via β -decay on a low background gas-flow proportional detector with anticoincidence circuitry (G542 System, Gamma Products), then for 207 Bi via γ -emission to determine yield. Polonium was then removed from the SPE disks, plated to copper, and α -counted for 210 Po and the yield monitor 209 Po. Reported activities (±1 SD) include propagated uncertainties in the decay rate, detector efficiencies, and yield. Activity calculations and corrections are fully described by Waples (2020).

3. Results and discussion

Dissolved, particle-bound and total in situ activities of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in rain and water from the Milwaukee Inner Harbor in Lake Michigan are presented in Table 1 and Fig. 2. Comparisons between the activities reported here to other measurements of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in the literature are given by Waples (2020).

3.1. ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activity in rain

A total of 5.39 L of rainwater was collected and split into two samples for replicate analyses (Rain A and Rain B; Table 1). Mean total activities (\pm SD of propagated counting errors) of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in rain, decay corrected to the end of the rain event at 09:00 on 05 November, were 17,000 \pm 300, 8200 \pm 300, and 870 \pm 50 dpm m⁻³, respectively. Rainfall over the 40-h collection period was 2.49 cm. This translated to an atmospheric deposition of 423 \pm 8 dpm ²¹⁰Pb m⁻², 204 \pm 7 dpm ²¹⁰Bi m⁻², and 22 \pm 1 dpm ²¹⁰Po m⁻² at the end of the rain event (Fig. 2a). Using an average slip depth of 4 m, the rainwater itself would have elevated well-mixed water activities of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in the slip by 106, 51, and 6 dpm m⁻³, respectively. Surface runoff and drainage (of rain and particulate matter) into the slip would have added considerably more activity.

No additional precipitation was recorded at the study site for the duration of the lake water sampling period (08–20 November), and it was assumed that dry deposition of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po was negligible during that time.

Table 1

Activities (±1SD) of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in rain and lake water samples.

• •	, ,		1			
Sample	Type ^a Collected date (201		Δt^{b} days	210 Pb dpm m $^{-3}$	210 Bi dpm m $^{-3}$	210 Po dpm m $^{-3}$
rain A	total	11–5	0.0	$16{,}600\pm600$	8300 ± 300	850 ± 40
rain B	total	11–5	0.0	$17{,}300\pm300$	8000 ± 500	890 ± 90
water (LS7P)	part.	11-8	3.0	207 ± 8	198 ± 10	89 ± 3
water (LS7D)	diss.	11-8	3.0	$141\pm 32^{\circ}$	199 ± 10	55 ± 2
water (LS8T)	total	11-8	3.0	347 ± 31	397 ± 14	144 ± 4
water (LS9P)	part.	11–15	10.1	38 ± 3	49 ± 3	22 ± 1
water (LS10P)	part.	11–15	10.1	41 ± 4	47 ± 4	21 ± 1
water (LS9D)	diss.	11–15	10.1	87 ± 7	79 ± 7	36 ± 2
water (LS10D)	diss.	11–15	10.1	85 ± 8	90 ± 8	33 ± 1
water (LS11T)	total	11–15	10.1	125 ± 7	133 ± 12	56 ± 3
water (LS12P)	part.	11–20	15.1	51 ± 3	51 ± 4	34 ± 1
water (LS13P)	part.	11–20	15.1	47 ± 3	45 ± 4	32 ± 1
water (LS12D)	diss.	11–20	15.1	54 ± 8	33 ± 5	25 ± 1
water (LS13D)	diss.	11–20	15.1	58 ± 7	46 ± 4	25 ± 1
water (LS14T)	total	11-20	15.1	106 ± 7	88 ± 9	58 ± 3
water (LS15T)	total	11–20	15.1	105 ± 7		

^a total = total sample; part. = particulate fraction; diss. = dissolved fraction ($< 0.45 \mu m$).

^b Days since end of rain event on 11/5/2018.

^c Italicized values calculated by difference or sum (errors propagated).

3.2. ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activity in lake water

Total, particle-bound, and dissolved (< 0.45 μm) activities of $^{210}\text{Pb},$ $^{210}\text{Bi},$ and ^{210}Po in lake water on 08, 15, and 20 November are shown in Table 1.

Total ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activities fell steeply from their initial values on 08 November (~ 3 days after the rain event) to 15 November, and ²¹⁰Pb and ²¹⁰Bi activities continued to decrease at a slower rate from 15 November to 20 November, while ²¹⁰Po activity increased slightly (Fig. 2a). Total ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po nuclide activity ratios ranged from 1:1.14:0.41 on 08 November to 1:1.06:0.44 on 15 November to 1:0.83:0.55 on 20 November (Table 2). This is the first observation of a total ²¹⁰Bi/²¹⁰Pb activity ratio of less than one in an aquatic system.

The SPM concentration on 08 November was 3.62 g m^{-3} (Fig. 2c; Table 3). By 15 November, the SPM concentration had fallen to 0.64 g m⁻³. An increase in the SPM concentration to 1.26 g m⁻³ by 20 November suggested that a resuspension event had taken place over the sampling interval.

Particle-bound ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activities trended with SPM concentrations (Fig. 2c), with highest activities per unit particle mass occurring when SPM concentrations were at their lowest on November 15 (Table 3). Particle-bound ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activity ratios ranged from 1:0.96:0.43 on 08 November to 1:1.22:0.54 on 15 November to 1:0.97:0.67 on 20 November (Table 2). Notable trends in activity ratios included: the temporary increase in the ²¹⁰Bi/²¹⁰Pb activity ratio from \sim unity to 1.22 ± 0.04 on November 15, the increase in the ²¹⁰Po/²¹⁰Bi activity ratio after the resuspension event on November 20, and the steady increase in the ${}^{210}Po/{}^{210}Pb$ activity ratio over the course of sampling to a maximum ratio of 0.67 \pm 0.04 on November 20. Particulate 210 Po/ 210 Pb activity ratios <1 have been found in coastal zones around the world (Tang et al., 2018), which indicates the importance of both atmospheric nuclide inputs [with ²¹⁰Po/²¹⁰Pb activity ratios $\ll 1$] and enhanced scavenging and removal in shallow water systems.

Dissolved nuclide activity fractions fell steadily from their initial measurement on 08 November to 20 November (Fig. 2b). Dissolved 210 Pb, 210 Pb, and 210 Po activity ratios ranged from 1:1.42:0.39 on 08 November to 1:0.99:0.40 on 15 November to 1:0.71:0.44 on 20 November, where – over the course of sampling – 210 Bi/ 210 Pb activity ratios decreased by half, 210 Po/ 210 Bi activity ratios doubled, and 210 Po/ 210 Pb activity ratios remained nearly constant (Table 2). The 210 Bi/ 210 Pb activity ratio > 1 on the dissolved nuclide fraction on 08 November is interesting. I speculate that: (1) either the dissolved 210 Pb activity [calculated as the difference between total and particle-bound

²¹⁰Pb measurements] on 08 November is wrong, or (2) ²¹⁰Pb was preferentially scavenged prior to the 08 November sampling date due to differences in the response of all three nuclides to particle concentration effects (Benoit and Rozan, 1999), or (3) ²¹⁰Bi was preferentially transported to the slip from the surrounding watershed during the rain event. A plausible mechanistic explanation for the latter hypothesis is given below in the *Radionuclide fluxes* section.

Distribution coefficients (K_d , L kg⁻¹) for ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po over the three sampling dates are shown in Fig. 3. Lower K_d values on 08 November were clearly influenced by higher SPM concentrations and the recent addition of large dissolved nuclide fractions in precipitation. For samples collected on 15 and 20 November, the geometric means of K_d values for ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po were 7.1 (±0.4) × 10⁵, 9.3 (±1.7) × 10⁵, and 10.2 (±0.6) × 10⁵ L kg¹, respectively. These ²¹⁰Pb and ²¹⁰Po K_d values are within the range of what has been found in other freshwater systems (surveyed by Mudbidre et al., 2014) and are also typical of nearshore marine systems. For instance, in the nearshore Mediterranean Sea study by Tateda et al. (2003), where SPM concentrations ranged from 1.4—5.0 g m⁻³, K_d values for ²¹⁰Pb and ²¹⁰Po ranged from ~0.2—1.5 × 10⁵ and ~ 0.1—6.8 × 10⁵ L kg⁻¹, respectively.

The ratio of the K_d values – or fractionation factor (F) – gives some indication of the degree of preferential sorption of one nuclide over another to particles (e.g., Mudbidre et al., 2014; Tang et al., 2017). For the sampling dates of 15 and 20 November, K_d values for Po > Bi > Pb, with $F_{Bi/Pb}$ values of 1.2 and 1.4, $F_{Po/Bi}$ values of 1.1 and 1.1, and $F_{Po/Pb}$ values of 1.4 and 1.5.

on each respective date. By comparison, $F_{Po/Pb}$ values ranged from 0.1—2.3 in the Clinton River (Mudbidre et al., 2014), 0.2—6.4 in the Yellow Sea (Hong et al., 1999), and averaged 1.6 along the North Atlantic GEOTRACES GA01 transect (Tang et al., 2018).

The only other measurement of both dissolved and particle-bound fractions of ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in an aquatic system – consisting of one sample from the Irish Sea (Biggin et al., 2002) –gives $F_{Bi/Pb}$ and $F_{Po/Pb}$ values of 20.6 and 10.7, with an $F_{Po/Bi}$ value of 0.5.

3.3. Radionuclide fluxes

Because ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po are all particle-reactive, each of the nuclides can, in theory, be used to trace particle flux in the water column. In shallow aquatic systems, however, a number of factors have to be considered including: (1) prevailing non-steady-state (NSS) conditions $(\partial A/\partial t \neq 0)$; e.g., Waples and Klump, 2013), which can apply to both the daughter and parent for the ²¹⁰Bi/²¹⁰Pb pair; (2) the proximity of the bottom and the effects of bottom scavenging on radionuclide



Table 2

Nuclide activity	ratios	for total,	particulate,	and	dissolved	(< 0.45	μm)	sample
fractions.								

Date (2018)		²¹⁰ Bi/ ²¹⁰ Pb	²¹⁰ Po/ ²¹⁰ Bi	²¹⁰ Po/ ²¹⁰ Pb
11-8		1.14 ± 0.11	0.36 ± 0.02	0.41 ± 0.04
11–15		1.06 ± 0.11	$\textbf{0.42} \pm \textbf{0.04}$	$\textbf{0.44} \pm \textbf{0.03}$
11-20	Total	$\textbf{0.83} \pm \textbf{0.08}$	$\textbf{0.66} \pm \textbf{0.07}$	0.55 ± 0.02
11-8		$\textbf{0.96} \pm \textbf{0.06}$	$\textbf{0.45} \pm \textbf{0.03}$	$\textbf{0.43} \pm \textbf{0.02}$
11–15		1.22 ± 0.04	$\textbf{0.45} \pm \textbf{0.01}$	0.54 ± 0.02
11-20	Particulate	$\textbf{0.97} \pm \textbf{0.07}$	0.69 ± 0.05	$\textbf{0.67} \pm \textbf{0.04}$
11-8		1.42 ± 0.33	$\textbf{0.28} \pm \textbf{0.02}$	0.39 ± 0.09
11–15		$\textbf{0.99} \pm \textbf{0.07}$	$\textbf{0.40} \pm \textbf{0.03}$	$\textbf{0.40} \pm \textbf{0.02}$
11-20	Dissolved	0.71 ± 0.11	0.63 ± 0.10	$\textbf{0.44} \pm \textbf{0.02}$

Ratio errors are propagated counting errors (± 1 SD) or \pm MAD for all duplicate sample means.

Table 3 Activities (± 1 SD) of 210 Pb, 210 Bi, and 210 Po on particles.

Sample	Collected date (2018)	SPM g m ⁻³	²¹⁰ Pb dpm g ⁻¹	²¹⁰ Bi dpm g ⁻¹	²¹⁰ Po dpm g ⁻¹
SPM (LS7P)	11–8	3.6	57 ± 2	55 ± 3	25 ± 1
SPM (LS9P)	11–15	0.6	60 ± 5	77 ± 5	34 ± 1
SPM (LS10P)	11–15		64 ± 6	74 ± 6	33 ± 1
SPM (LS12P)	11–20	1.3	41 ± 3	40 ± 3	27 ± 1
SPM (LS13P)	11–20		38 ± 3	36 ± 3	25 ± 1



Fig. 2. ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po activities in lake water from the harbor slip. a) Total nuclide activities; b) dissolved nuclide activities; c) particle-bound nuclide activities and measured SPM concentrations. Error bars show propagated counting errors (± 1 SD) or \pm MAD for all duplicate sample means. Timing of rain event is shown in panel a.

Fig. 3. Partition coefficients (K_d) for ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po in lake water samples. Error bars show propagated counting errors (± 1 SD) for 08 November measurements and propagated MAD errors for all other values calculated from duplicate sample means.

activity gradients (e.g., Craig et al., 1973); (3) transport phenomena and the relative importance of vertical turbulent diffusion to that of gravitational sinking (e.g., Lick, 1982); and (4) the resuspension of particle-bound nuclides back into the water column.

A mass balance for 210 Pb, 210 Bi, and 210 Po – from the surface to some specified depth (*z*) – can be expressed as:

$$\frac{\partial A_D}{\partial t} = \lambda (A_P - A_D) - J_{A_D} + V + I + R \tag{1}$$

where $\partial A_D / \partial t$ is the change in the daughter nuclide activity (A_D) of ²¹⁰Pb, ²¹⁰Pb, ²¹⁰Po over time, A_P is the [respective] total parent nuclide activity of ²²⁶Ra, ²¹⁰Pb, or ²¹⁰Bi, λ is the decay constant of the daughter nuclide, J_{A_D} is the removal flux of the daughter nuclide (e.g., Savoye et al., 2006; Tang et al., 2019), *V* is the sum of all advective and diffusive fluxes, *I* is the atmospheric flux of the daughter nuclide, and *R* is the resuspension of the daughter nuclide back into the water column.

Not all of the terms in Eq. (1) can be addressed with the data set presented here. However, a simple mass balance of the input and removal of 210 Pb, 210 Bi, and 210 Po and the change in SPM over time reveals several interesting processes (Table 4). The first three rows of Table 4 show the measured values of SPM and nuclide activities used to calculate the subsequent removal terms, as well as the observed mass/ activity ratios for each of the three nuclides.

Because air temperatures over the sampling period ranged from -8 °C to +7 °C, I could reasonably assume the 4-m water column was no longer thermally stratified and therefore well mixed. I could then normalize the removal terms for total, dissolved, and particle-bound nuclide fractions to a water column of 1 m depth. The right-hand-side

$$\frac{\partial A_D}{\partial t} = \frac{A_{D(t2)} - A_{D(t1)}}{t_2 - t_1}.$$
(2)

The net growth of the daughter nuclide $(\lambda(A_P - A_D))$ over each time interval was calculated as:

$$\lambda(A_P - A_D) = \lambda \left(\frac{A_{P(t1)} + A_{P(t2)}}{2} - \frac{A_{D(t1)} + A_{D(t2)}}{2} \right),\tag{3}$$

using the daughter nuclide decay constants of $\lambda_{Pb} = 8.510 \times 10^{-5} \text{ d}^{-1}$, $\lambda_{Bi} = 0.13835 \text{ d}^{-1}$, and $\lambda_{Po} = 0.0050 \text{ d}^{-1}$. The net ingrowth of ²¹⁰Pb was calculated using ²²⁶Ra as the parent, with an average Lake Michigan activity of 8 dpm m⁻³ (Rutgers van der Loeff et al., 2006). Atmospheric inputs (*I*) via dry deposition were assumed to be negligible over the course of the water sample collecting period and transport via advection and turbulent diffusion (*V*) were also assumed to be negligible for the purposes of this demonstration. Sediment resuspension (*R*) is discussed in further detail below.

Combining Eqs. (2) and (3), the flux for each daughter nuclide fraction (J_{A_p}) was calculated as:

$$J_{A_D} = \lambda \left(\frac{A_{P(t1)} + A_{P(t2)}}{2} - \frac{A_{D(t1)} + A_{D(t2)}}{2} \right) - \frac{A_{D(t2)} - A_{D(t1)}}{t_2 - t_1}.$$
 (4)

It should be noted that the approximated calculation of J_{AD} shown in Eq. (4), follows the same strategy given by Buesseler et al. (1992) and is most sensitive – in terms of increased error – for the shortest-lived nuclide, ²¹⁰Bi. However, a rigorous calculation of J_{Bi} (from Friedrich and Rutgers van der Loeff, 2002), where:

$$J_{Bi} = \lambda_{Bi} \left(\frac{\frac{\lambda_{Pb}A_{Ba} - J_{Pb}}{\lambda_{Pb}} \left[\frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \left(e^{-\lambda_{Bi}t} - e^{-\lambda_{Pb}t} \right) + \left(1 - e^{-\lambda_{Bi}t} \right) \right] + A_{Pb(t1)} \frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \left(e^{-\lambda_{Pb}t} - e^{-\lambda_{Bi}t} \right) + A_{Bi(t1)} e^{-\lambda_{Bi}t} - A_{Bi(t2)}}{1 - e^{-\lambda_{Bi}t}} \right), \tag{5}$$

of Eqs. (2)–(5), therefore, implicitly include a depth term factor of 1 m and can be expressed in units of dpm $m^{-2}\,d^{-1}.$

The first removal term begins with the observed change in activity over time $(\partial A_D/\partial t)$, which was calculated as:

shows the approximation introduces an insignificant mean error of 0.3 \pm 0.2 dpm m⁻² d⁻¹ for all dissolved, particulate, and total nuclide fractions. Similar calculations for ^{210}Po result in a smaller mean flux error of 0.03 \pm 0.02 dpm m⁻² d⁻¹. These errors would decrease as Δt gets smaller.

Table 4

A mass balance of ²¹⁰Pb, ²¹⁰Bi, ²¹⁰Po, and suspended particulate matter removal from the water column.

Date	Time days	$\rm SPM~g~m^{-3}$	Total			Dissolved			Particulate			Mass/activity		
			²¹⁰ Pb	²¹⁰ Bi	²¹⁰ Po	²¹⁰ Pb	²¹⁰ Bi	²¹⁰ Po	²¹⁰ Pb	²¹⁰ Bi	²¹⁰ Po	²¹⁰ Pb	²¹⁰ Bi	²¹⁰ Po
			dpm m ⁻³			dpm m ⁻³			dpm m ⁻³			$g dpm^{-1}$		
08-Nov	0.00	3.62	347	397	144	141	199	55	207	198	89	0.017	0.018	0.041
15-Nov	7.03	0.64	125	133	56	86	85	34	39	48	21	0.016	0.013	0.030
20-Nov	12.03	1.26	105	88	58	56	40	25	49	48	33	0.026	0.026	0.038
	Time interval		dpm m ⁻	$dpm \ m^{-2} \ d^{-1}$		$dpm \ m^{-2} \ d^{-1}$		$dpm \ m^{-2} \ d^{-1}$						
dA/dt	1		-31.6	-37.6	-12.6	-7.8	-16.3	-3.0	-23.8	-21.3	-9.6			
	2		-4.0	-9.0	0.5	-5.9	-9.0	-1.8	2.0	0.0	2.3			
$\lambda(A_{P}-A_{D})$	1		0.0	-3.9	0.8	0.0	-4.0	0.5	0.0	0.0	0.3			
	2		0.0	0.7	0.3	0.0	1.2	0.2	0.0	-0.5	0.1			
Nuclide removal	1		31.6	33.6	13.4	7.8	12.3	3.5	23.8	21.3	9.9			
	2		4.0	9.7	-0.2	5.9	10.2	2.0	-2.0	-0.5	-2.2			
Mass/activity (calc)	1											0.018	0.020	0.043
	2											0.063	0.268	0.057
										$g m^{-2} d^{-1}$				
Mass removal (calc)	1								0.42	0.42	0.42			
	2								-0.12	-0.12	-0.12			

Time interval 1: 08-15 November; time interval 2: 15-20 November.

The flux estimates in Table 4 show that during the first time interval (08—15 November), when SPM concentrations fell by 82%, particlebound nuclide fluxes ($J_{AD^{per}}$) of 210 Pb, 210 Bi, and 210 Po accounted for 74%, 62%, and 74% of each nuclide's respective total nuclide flux ($J_{AD^{per}}$), with the remaining drawdown occurring in the dissolved nuclide fractions. During the second time interval (15—20 November), when SPM concentrations increased by 97%, essentially 100% of the net loss of total nuclide activity was due to the removal of the dissolved nuclide fractions – presumably by direct sorption to bottom sediments. We have made similar observations of a significant removal of the dissolved nuclide fraction by bottom scavenging in the Milwaukee Outer Harbor using dual tracers of ⁷Be and 131 I (Montenero et al., 2017), and in nearshore Lake Michigan using dual tracers of 234 Th and 90 Y (Waples, 2015; Waples et al., 2017).

The observed [net] fluxes of SPM (J_{mass}) during the first (08—15 November) and second (15—20 November) time intervals were 0.42 and - 0.12 g m⁻² d⁻¹, respectively (with the negative sign indicating an increase in water column concentration). Assuming SPM was primarily mineral in composition and conservative by nature, a mass balance calculation of the mass/activity ratios ($mass/A_{Dpart}$) of particles leaving (time interval 1) and re-entering (time interval 2) the water column was determined as:

$$\frac{mass}{A_D^{part}} = \frac{J_{mass}}{J_{A_D^{part}}}.$$
(6)

The calculated mass/activity ratios (Table 4) for the first time interval were 2%, 10%, and 5% higher than the observed ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po mass/activity ratios measured on 08 November, and suggested the removal of larger particles (with higher mass/activity ratios), leaving smaller particles with lower mass/activity ratios in the water column on 15 November. Also interesting are the very high calculated mass/activity ratios of resuspended particles during the second time interval. The corresponding [calculated] particle activity/mass ratios for ²¹⁰Pb, ²¹⁰Bi, and ²¹⁰Po were 15.8, 3.7, and 17.7 dpm g⁻¹, respectively. The approximate equilibrium in activity between ²¹⁰Pb and its granddaughter ²¹⁰Po suggests older sediments were mobilized into the water column, but the low calculated activity of ²¹⁰Bi is surprising (see Table 3 for comparison).

An interesting and plausible mechanistic explanation for both the low ²¹⁰Bi activity on resuspended sediment and the dissolved ²¹⁰Bi/²¹⁰Pb activity ratio > 1 that was observed just after the rain event on 08 November is given by Ulrich and Degueldre (1993). They found that when ²¹⁰Bi in the dissolved phase sorbed to a particle of montmorillonite (clay), the sorption was nearly irreversible. However, if ²¹⁰Pb sorbed to a particle and subsequently decayed to ²¹⁰Bi, hot atom chemical effects (Wolfgang, 1965) and differences in preferential sorption sites between ²¹⁰Pb and ²¹⁰Bi led to a decrease in ²¹⁰Bi K_d of two to three orders of magnitude. If desorbed ²¹⁰Bi then subsequently sorbed to colloidal (i.e., dissolved fraction) material, the unexpected distributions of ²¹⁰Bi found here might be valid. This remains as speculation until direct analysis of resuspended sediment and rainwater runoff are made.

3.4. The use of ²¹⁰Bi as a particle tracer in high flux environments

The use of any radionuclide as a particle tracer depends most importantly on the nuclide's affinity for particle sorption and its half-life in relation to the timescale of the scavenging process. This can be demonstrated using the four daughter/parent [grandparent] nuclide pairs of $^{210}Po/[^{210}Pb]$, $^{234}Th/^{238}U$, $^{210}Bi/^{210}Pb$, and $^{90}Y/^{90}Sr$ (Fig. 4). In a 1-D steady-state (SS) system with low particle flux (Fig. 4, left panel), ^{210}Po (t½: 138.4 days) and ^{234}Th (t½: 24.1 days) will show significant disequilibria with ^{210}Pb and ^{238}U , respectively, while short-lived nuclides of ^{210}Bi (t½: 5.01 days) and ^{90}Y (t½: 2.67 days) remain in near equilibrium with their parents (^{210}Pb and ^{200}Sr , respectively). Moreover, the slope of change in disequilibria with increasing particle flux is very low with the short-lived nuclides [^{90}Y and ^{210}Bi] so that small uncertainties in measured daughter/parent disequilibria translate to relatively large uncertainties in particle flux. In high flux environments,



Fig. 4. Estimated daughter/parent nuclide activity ratios in offshore (left) and nearshore (right) water columns as a function of the 234 Th removal rate on particles in a 1-D SS system. All nuclide pairs scaled to 234 Th removal using K_d values determined by Waples (2015) and this study. Site specific K_d values will alter these relationships. 238 U and 90 Sr were assumed to behave conservatively. The 234 Th removal rate in each cited study was calculated as the removal flux of 234 Th divided by the inventory of 234 Th over the stated water column depth (Buesseler et al., 2008; Cochran et al., 2000; Tanaka et al., 1983; Gustafsson et al., 1998; McKee et al., 1984). Vertical diffusive fluxes of 234 Th in nearshore Lake Michigan were assigned to particle flux in this exercise.

however, the opposite is true (Fig. 4, right panel). Here, the longer-lived nuclides [²¹⁰Po and ²³⁴Th] are scavenged to such an extent that they are difficult to measure, and small uncertainties in measured daughter/ parent disequilibria again translate to large uncertainties in particle flux.

The use of ⁹⁰Y as a particle tracer in a high flux, nearshore water column has been demonstrated in Lake Michigan (Waples, 2015; Waples et al., 2017). However, its practical use as a tracer is limited to the few systems where ⁹⁰Sr activities are high (e.g., the Laurentian Great Lakes and the Irish Sea; Waples and Orlandini, 2010). The same can be said of ²³⁴Th, where rapid drawdown of ²³⁴Th activity in the water column (e. g., Waples and Orlandini, 2010; Waples and Klump, 2013) or low activities of ²³⁸U (e.g., ²³⁸U activity in Lake Superior is 0.04 dpm L⁻¹; J. T. Waples, unpubl.) can render the use of this tracer impractical.

Because the distribution of ²¹⁰Pb is global and the half-life of ²¹⁰Bi is short, the data presented here support the use of ²¹⁰Bi as a tracer of particulate matter in nearly any environment where rapid scavenging and the preferential removal of ²¹⁰Bi leads to significant ²¹⁰Bi/²¹⁰Pb disequilibrium (Fig. 4). Using any naturally occurring radionuclide as a tracer in a shallow dynamic system, however, is a nontrivial exercise. Non-steady state conditions prevail as a rule and all terms in Eq. (1) must be considered. This significant effort is, nevertheless, necessary if we are to make progress in our empirical understanding of particle dynamics and chemical cycling in lakes and shallow marine systems.

Declaration of Competing Interest

None.

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